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# Extending the area of investigation of fine versus coarse quartz optical ages from the Lower Danube to the Carpathian Basin

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## Abstract

Despite the general satisfactory performance of quartz in the single aliquot regeneration protocol (SAR), previous optically stimulated luminescence (OSL) dating studies of key loess sections in Romania, Lower Danube region, revealed a disturbing disagreement among the ages obtained on fine (4-11µm) grains and coarse (63-90µm) grains respectively. The current study aims at expanding these investigations, both by extending the area of study from the Lower Danube Basin to the Carpathian Basin and by applying time-resolved optically stimulated luminescence (TR-OSL) on quartz, in order to gain further insights into the above mentioned behaviour. The samples from Orlovat loess paleosol section (Vojvodina, Serbia) showed a similar behaviour to that previously reported on Romanian loess. A marked difference between the dose saturation characteristics of fine and coarse quartz OSL signals is observed for both continuous wave (CW-OSL) and pulsed OSL (POSL), where the dose response (up to 1000 Gy) is well described by a sum of two saturating exponential functions. TR-OSL measurements show one single, characteristic quartz lifetime for both natural as well as regenerative signals in the entire dose range investigated. A general disagreement between the ages obtained on the two grain sizes for samples with equivalent doses higher than about 100 Gy is reported as in the case of Romanian loess, inferring that the age discrepancy between the two grain sizes might be more widespread than previously thought.

**Keywords:** quartz, loess, single aliquot regeneration (SAR) protocol, dose response, time-resolved optically stimulated luminescence (TR-OSL).

## 1. Introduction

The loess paleosol sequences of the Carpathian Basin and Lower Danube region (Serbia, Romania, Bulgaria) are thought to represent the most continuous and high resolution archives of regional climate and environmental change during the Late and Middle Pleistocene, (Fitzsimmons et al., 2012), and a link between European and Asian loess deposits (Marković et al., 2012). Their significance however, can only be fully understood only once a reliable and absolute chronology is available.

Technological developments in the past years have resulted in significant improvements in the achievable precision and accuracy using luminescence dating; it is now considered one of the most important chronometric methods in the study of the late Quaternary (Wintle, 2008). However, dating applications recently performed on Romanian loess by the application of optically stimulated luminescence on quartz lead to unexpected results.

A first study has reported optically stimulated luminescence (OSL) ages for the loess sequence near Mircea-Vodă (Dobrogea, SE Romania) using silt-sized (4-11 $\mu$ m) quartz as dosimeter (Timar et al., 2010). An internally consistent set of optical ages was obtained; however, a comparison of these ages with a magnetic time-depth model based on magnetic susceptibility measurements suggested a systematic underestimation beyond the penultimate glacial period (the SAR OSL ages of the three samples below the S1 soil were interpreted as age underestimates). Interestingly, the OSL signals from these samples did not indicate any odd characteristics: the dominance of the fast component in the OSL signals was indicated by the decay shape of the CW-OSL and LM-OSL signals; and the samples passed the procedural tests of the single-aliquot regenerative-dose (SAR) protocol (i.e. recycling ratio, recuperation, dose recovery and preheat plateau tests) indicating that the protocol should provide reliable results, while the natural signals were found well below the saturation region of the laboratory dose response.

It was concluded that optical dating of fine-grained quartz can be used to establish a reliable chronology for Romanian loess up to ~70 ka corresponding to an equivalent dose of ~ 200 Gy. Such a behaviour is consistent with results from old (>70 ka) Chinese loess (Buylaert et al., 2007), and with the more general suggestion that SAR may underestimate the true age in the older age range (Murray et al., 2007; Lowick et al. 2010a,b, 2011). Thus, apparently reliable OSL laboratory measurement procedure does not necessarily guarantee an accurate

determination of the true burial dose. A subsequent study of coarse-grained (63-90  $\mu\text{m}$ ) quartz extracted from the same section (**Timar-Gabor et al., 2011**) revealed that equivalent doses obtained for coarse quartz grains were systematically larger than those for the fine grains; the observed difference was too high to be explained by partial bleaching or microdosimetric effects. Furthermore, both quartz fractions passed the procedural tests of the single-aliquot regenerative-dose (SAR) protocol and yielded an internally consistent set of optical ages.

**Timar-Gabor et al. (2012)** investigated into the shape of the dose response curve for the two grain size fractions in the high dose region of 5-10 kGy; they observed an age discrepancy in the Mostiștea section (Romanian plain) similar to that in the Mircea-Vodă. Work on Costinești section (Dobrogea, SE Romania) (**Constantin et al., 2014**) again confirms the same trend. From the study of **Timar-Gabor et al. (2012)** two additional issues of general importance to the SAR protocol emerged. Firstly the natural signal of an infinitely old sample was found not to be in saturation, perhaps implying that the dose response measured in the laboratory may not simulate trapped charge growth during burial; this aspect was further investigated by **Timar-Gabor and Wintle (2013)**. Secondly, it was observed that the dose response curve for coarse grains (63-90  $\mu\text{m}$ ) is very different from fine grains (4-11  $\mu\text{m}$ ), the latter saturating at much higher doses. A similar observation has also been made by **Constantin et al. (2012)** where dose response curves of fine grains (4-11  $\mu\text{m}$ ) and coarse grains of different sizes (63-90, 90-125, 125-180  $\mu\text{m}$ ) were compared for Căciulătești site (SW Romania). This different dose response pattern was also reported for quartz extracted from loess in Western Europe by **Kreutzer et al. (2012)**. This is mostly intriguing in correlation to the fact that the fine fraction underestimates the true ages earlier than the coarse fraction does. Also, it is important to note that the values obtained for the saturation characteristics for the fine quartz fraction are very similar to the values reported by other international studies (e.g. **Lowick et al., 2010b**) in their work on fine material, while the saturation characteristic doses obtained in this study for coarse grained quartz are close to the values previously reported by others on coarse material from other locations (e.g. **Murray et al., 2007; Pawley et al., 2010**).

In a geochemical characterisation study performed by **Buggle et al. (2008)** it was confirmed that both Serbian and Dobrudjan loess have a major component derived from Danube alluvium. As second material sources, loess in the Dobrudja region showed a significant contribution of a second loess source, probably the glaciofluvial sediments of the Ukraine, to which variable

contributions from local sand dune fields can be considered. Although the different origin of the sedimentary grains in loess should not justify the difference in equivalent doses that leads to a systematic age offset between the coarse and fine grains ages, it should be taken into consideration whether the different properties of fine and coarse grains can be due to their possible different origin. In the present study we are extending the investigated area by analysing whether the above mentioned finds are more than a local feature of the eastern part of the Lower Danube basin or do apply to the Carpathian Basin loess as well.

## 2. Samples, instrumentation and measurement protocol

The present work focuses on eight loess samples collected from a loess-paleosol sequence exposed in a brickyard at the village of Orlovat, located in the Tamiš loess plateau (**Figure 1**). Out of these, samples ORL-8 to ORL-4 have been collected from L1 unit, samples ORL 3 and ORL 2 have been sampled from S1 unit, while sample ORL 1 belongs to L2 unit, thus a total age span of more than 130 ka being covered (**Figure 2**). The importance of the Orlovat section as a key archive for the late Pleistocene paleoclimate and paleoenvironment of the southeastern part of the Carpathian Basin has been emphasized by previous multi-proxi studies (**Marković et al., 2014; Lukić et al. 2014**) to which the reader is referred to for more information.

OSL dating of coarse (63-90µm) quartz was also documented in **Marković et al. (2014)**. Fine quartz extraction followed the conventional procedures described in our previous studies (**Timar et al. 2010; Timar-Gabor et al. 2011; Constantin et al., 2014**). All continuous wave optically stimulated luminescence measurements (CW-OSL) were made in the Cluj Luminescence Dating Laboratory with a Risø TL/OSL DA-20 reader equipped with blue light emitting diodes (LEDs) emitting at  $470 \pm 30$  nm and IR LEDs emitting at  $875 \pm 80$  nm; luminescence signals were observed through a 7.5 mm thick Hoya U-340 UV filter. Irradiations have been carried out using a  $^{90}\text{Sr}$ - $^{90}\text{Y}$  beta source, calibrated using fine and coarse quartz supplied by Risø National Laboratory. The dose rate, for the time of measurement, determined for coarse grains mounted on stainless steel disks was 0.147 Gy/s, while the dose rate for fine grains deposited on aluminium disks amounted to 0.118 Gy/s. Details of the measurement apparatus can be found in **Thomsen et al. (2006)**. The OSL signal was collected in time intervals of 0.154 s. All samples have been analysed in a SAR protocol (**Murray and Wintle, 2000, 2003**). The OSL signal used

for analysis was that obtained for the first 0.308 s of the decay curve minus a background derived from the signal measured between 2.464 and 3.080 s, as had been used in previous studies on Romanian loess (Timar et al., 2010; Timar-Gabor et al., 2011; Timar-Gabor et al., 2012; Timar-Gabor and Wintle, 2013) and the recommendations of Cunningham and Wallinga, (2010). Natural and regenerated signals were measured after a preheat of 10 s at 220°C unless otherwise stated; the response to the test dose (16 Gy) was measured after a cutheat to 180°C. The value of the test dose was kept constant through all measurements for both grain sizes of quartz. After the measurement of the response to the test dose, a high-temperature bleach was performed by stimulating with the blue diodes for 40 s at 280°C (Murray and Wintle, 2003). Time resolved optically stimulated luminescence (TR-OSL) experiments have been carried out in Risø National Laboratory on a Risø TL/OSL-20 (their reader V) equipped with an integrated pulsing option to control the LEDs, and a Photon Timer attachment to record the TR-OSL described in detail in Lapp et al. (2009). All TR-OSL experiments have been carried out using the same measurement parameters previously used in CW-OSL. For stimulating and recording the TR-OSL signals the total measurement time was set to 100 s, with a pulse period of 500 µs consisting of an on time of 50 µs and an off time of 450 µs; this can be translated to a net stimulation period equivalent to 10s in CW-OSL. A total of 500 data points were used for data collection in pulsed stimulation out of which the first and the last five channels were dead channels. Measurement of dose response curves using pulsed optically stimulated luminescence measurements have been specially designed in order to reproduce the CW-OSL measurements conditions as closely as possible, and to minimise any possible feldspar contamination to quartz, by: (i) selection of an on time of 50 µs (ii) gating the photomultiplier for counting only during the off period; (iii) ignoring the first 4µs during the off time in TR-OSL OSL data analysis (Ankjaergaard et al., 2010).

The specific activities of radionuclides of interest for dose rate determination ( $^{238}\text{U}$ -series,  $^{232}\text{Th}$  series and  $^{40}\text{K}$ ) were obtained through high-resolution gamma spectrometry using an ORTEC hyperpure germanium detector having the following characteristics: active volume of 181 cm<sup>3</sup>, 0.878 keV FWHM at 5.9 keV, 1.92 keV FWHM and 34.2% relative efficiency at 1332.5 keV, calibrated in efficiency using International Atomic Energy Agency standards.

### 3. Continuous wave optically stimulated luminescence investigations and age results

All samples investigated displayed bright and rapidly decaying OSL signals. Dose recovery tests have been carried out on every sample according to the methodology outlined by **Murray and Wintle (2003)**. The given dose was chosen to be as close as possible to the estimated equivalent dose and at least three aliquots have been used for each sample. **Figure 3** presents the results obtained for fine grains (panel a)) alongside with the results obtained on coarse grains (panel b)) previously presented by **Marković et al. (2014)**. It can be noted that for all samples laboratory given doses can be measured both accurately and precisely, although a slight trend towards overestimation of the given dose can be observed in both grain size fractions.

Equivalent doses have been measured using at least 12 aliquots in the case of coarse (63-90  $\mu\text{m}$ ) quartz except sample ORL 2 where analysis have been carried out on eight aliquots due to lack of material. For fine (4-11  $\mu\text{m}$ ) quartz, which show more reproducible results at least 7 aliquots have been used. **Table 1** summarizes all data relevant to equivalent dose measurement, while **Table 2** presents the relevant information for age determination. It can be noted that both grain size fractions show a general compliance with the prerequisites for the SAR protocol, such as recycling, IR depletion, recuperation and dose recovery. The age discrepancy between coarse and fine grains previously reported for Romanian loess, is also observed for the samples investigated in this study, except for the youngest two samples (ORL 8 and ORL7). As in the case of our previous investigations, the age difference can be attributed to the difference in equivalent doses; contrary to the dosimetric expectation, the values derived from the coarse quartz are systematically higher than the values obtained on fine quartz.

One possible cause of concern is related to a thermal instability of the OSL signal sampled from the fine fraction. This could be caused either by contamination of the signal with an unstable medium OSL component (see e.g. **Choi et al., 2003; Li and Li, 2006; Steffen et al., 2009**), or by a thermal instability of the fast OSL component itself (**Fan et al., 2011**). This has been tested using preheat plateau tests (**Figure 4a**) and high resolution pulse annealing experiments for both natural and regenerated signals (**Figure 4b**). There is no dependency of the measured equivalent dose on preheat temperature and the three datasets obtained in pulse anneal experiments (natural, regenerated and calibration quartz) suggest that the stability of the fast component for both grain sizes is the same, therefore, the long term stability of the signal should not be a problem.

One of the major differences in the behaviour of fine and coarse quartz grains previously reported for Romanian loess was observed in the different dose response saturation characteristics. We have examined this on the Orlovat samples by constructing dose response curves up to 1000 Gy using four different samples and at least three aliquots for each sample. The results are presented in **Figure 5**. As in the case of Romanian loess it can be observed that: (i) there is little scatter between aliquots of the same sample; (ii) the growth pattern for the same grain size is not age dependent; (iii) the growth pattern can be fitted well only with a sum of two exponential functions, (iv) although the dose response of the two grain sizes investigated has the same shape up to approximately 100 Gy, there is a marked difference between the fine and the coarse grain's saturation characteristics, with coarse grains saturating at much lower doses than the fine grains.

It is also interesting to note that samples ORL 7 and ORL 8 for which the OSL ages were found in agreement have both equivalent doses lower than 100 Gy, thus these values are in the region where the first exponential component is below saturation and where the dose response curves of the two grain sizes are consistent with each other. The same results have been obtained on a recent study on Romanian loess located in the southern Romanian Plain, in S-W Romania (**Constantin et al., online published**) where quartz ages obtained on fine and coarse quartz for the uppermost two young samples (ages about 11ka, and 20 ka respectively) were found in agreement.

#### **4. Time resolved optically stimulated investigations**

In pulsed stimulation (POSL) the incident photon flux is delivered with pulses with a certain pulse width, which is generically called the on-time. The stimulation pulses are separated by a period called off-time during which it is possible to measure the decay of the luminescence signal generated by the preceding stimulation pulse. The decay of this signal, registered as function of time on a nano to microscale range is called time resolved luminescence (TR-OSL). The shape of TR-OSL is mineral and trap dependent, and therefore it can be used as a powerful instrumental method for isolating quartz signals in mixed quartz-feldspars samples (**Denby et al., 2006; Thomsen et al., 2008**). Thus, by the application of pulsed stimulation one can confirm whether the growth of signal with dose observed in continuous wave stimulation is indeed a



characteristic of quartz, or is an experimental artefact caused by feldspar contamination that might have not been detected through IR depletion ratio tests.

On the other hand, the examination of the lifetimes derived from time resolved optically stimulated luminescence signals can give insights on the recombination processes in quartz. One possible mechanism for the different saturation characteristics observed for the signal dose response of the two grain sizes could be competition between different recombination centres. This competition may in turn be dependent on the different origins of fine and coarse quartz grains. This effect was investigated on sample ORL 4 on fine and coarse grains using TR-OSL. A typical quartz time resolved OSL curve recorded both during on and off time is presented in **Figure 6**.

Photon arrival time distributions have been recorded for natural signals as well as for different laboratory given doses ranging from 19 to 969 Gy, using a similar SAR measurement protocol as in the case of continuous wave experiments. During these experiments the photomultiplier was gated for counting only during the off period. The results obtained on one representative aliquot of fine grains are depicted in **Figure 7a**. It can be noted that during off time the signals display a slow decay, typical for quartz. It can be seen as well that a prior IR exposure of 100s at 125 °C does not influence the shape of the signal. This is further confirmation that the continuing growth of the signals up to very high doses noted in CW-OSL studies cannot be attributed to feldspar contamination. Similar results have been obtained on coarse quartz grains (data not shown here). The off-time photon-arrival-time distribution for all investigated doses can be fitted adequately with a single exponential function (see an example in **Figure 7b**).

The dose response curves constructed using pulsed OSL follow the same pattern as in the case of continuous wave OSL experiments (**Figure 8a**) showing beyond doubt that the discrepancy observed between coarse and fine grains is inherent in quartz OSL, and not due to some contaminant. Moreover, the equivalent doses obtained using pulsed OSL on four aliquots (ORL 4 fine  $De = 171 \pm 9$  Gy; ORL 4 coarse  $De = 170 \pm 16$  Gy) are in good agreement with the CW-OSL equivalent doses (see **Table 1**).

The average lifetime values for natural and regenerated signals obtained on coarse and fine grains are depicted in **Figure 8b** (four aliquots were used for each point). It can be observed that the values range in the typical interval for quartz (36.5-38.5 $\mu$ s) for a stimulation temperature of 125 °C (e.g. Chithambo, 2003; Ankjærgaard et al., 2010-figure 1; Pagonis et al., 2011) and

there is no marked dependence of lifetime as function of given dose, in contrast to the finds on certain samples of **Chithambo et al. (2008)**. Due to the strong thermal quenching observed for this emission, and the satisfactory application of the Mott–Seitz model to explain TR-OSL data (**Pagonis et al., 2010**) we can assume the likely possibility that this is a relaxation lifetime of the luminescence centre. Based on this we can deduce that it is likely that there is only one dominant centre emitting UV luminescence in our samples. In summary, our samples have the typical quartz recombination center which gives rise to the measured emission for all the given doses, for both fine and coarse grain samples.

## **5. Relation between stratigraphy and luminescence chronologies**

**Marković et al. (2014)** provided chronostratigraphic interpretation of the Orlovat section based on detailed litho- and pedo-stratigraphy, enviromagnetic parameters, as well as preliminary coarse grain quartz luminescence ages. The results show that the approximately 10 m thick section preserves an a typical Late Pleistocene succession for the Carpathian Basin (**Fitzsimmons et al., 2012**). Notably, the normally widespread pedocomplex V-L1S1 is missing (e.g. **Marković et al., 2008**). This contrasts with other parts of the sequence which appear extremely highly resolved, such as the thicker pedocomplex V-S1 and the detailed transitions between interglacial pedocomplexes V-S1 and V-S0, with the last glacial loess unit V-L1 (**Marković et al., 2014**).

**Figure 2** shows, similar to Romanian loess-paleosol sequences, that luminescence ages derived from coarser material (at least for equivalent doses higher than ~100 Gy) seem to better fit the expected geological ages. Samples taken from the penultimate glacial loess layer V-L2 and pedocomplex V-S1 obtained on coarse quartz provide an almost perfect match to the expected geological age.

At this section, evidence for the middle pleniglacial pedogenesis is not observed in contrast to other sections in the Vojvodina region (**Marković et al., 2008; Stevens et al., 2011**). This anomaly in the Orlovat enviromagnetic record indicates a potential depositional hiatuses between approximately 2.5 and 2 m depth in the Orlovat profile. This is partly supported by the luminescence dates. After a relatively smooth decrease in luminescence ages from the top of paleosol V-S1 to the middle part of the last glacial loess unit V-L1 a chronological shift occurs below and above sample ORL 6 indicating a significant reduction in accumulation rates, or the

likely occurrence of a hiatus in sedimentation (Marković et al., 2014). To identify more precisely these depositional gaps we plan to apply more detailed sampling for luminescence dating, similar to that presented in the recent studies of Stevens et al. (2006; 2007; 2008) on the Chinese Loess Plateau. Thus, two independent lines of evidence, the magnetic record and luminescence chronology, confirm a discontinuity in deposition as suggested by an incomplete stratigraphy when compared to sections further west. However, on the basis of the existing results it is very difficult to precisely indicate the age of the hiatuses at Orlovat loess-palaeosol sequence, especially because of the large variability in the sedimentological and pedological characteristics of pedocomplex V-L1S1 in the region (Marković et al., 2014).

Luminescence dates obtained on both coarse and fine material clearly demonstrate an Early Holocene age almost 0.7 m below the Holocene soil V-S0. This raises important questions regarding the onset of soil formation and the cessation of loess deposition in the region (Marković et al., 2014).

From a methodological point of view, it is very important to note the agreement between the fine and coarse quartz ages obtained for the two uppermost samples. The observed agreement can be considered an indication that the discrepancy between the fine and the coarse grain ages is a dose dependent phenomenon.

There is a clear better agreement of the coarse grained ages with the expected ages for the penultimate glacial loess layer V-L2 and pedocomplex V-S1. The coarse grain ages seem to provide a solid chronology for V-L1 as well, while the fine quartz ages seem to slightly underestimate the expected ages in the lower part of this unit. This was also observed for Romanian loess, but intriguingly, the application of post IR-IR<sub>225</sub> on polymineral fine grains on Mircea-Vodă section lead to obtaining feldspar ages that confirm the fine quartz chronology for L1, although in the case of L2 the agreement was observed between the post IR-IR<sub>225</sub> and coarse quartz ages (Vasiliniuc et al., 2012). Taking into consideration the above mentioned, as well as the fact that the cause of the observed discrepancy remains not understood none of the two chronologies can be discarded or favoured at the time being and further investigations are required.

## 6. Conclusions

Except for the two youngest samples, with equivalent doses of  $<100$  Gy, the remaining eight samples collected from L1, S1 and L2 units of Orlovat section (Vojvodina, Serbia) investigated in this work presented the same general OSL behaviour as quartz extracted from Romanian loess, meaning that the equivalent doses obtained on coarse quartz were systematically higher than the equivalent doses obtained using fine quartz. All samples passed adequately recycling, recuperation, IR depletion and dose recovery tests and the thermal stability of the investigated signals was confirmed through preheat plateau tests and pulse anneal experiments.

As in the case of quartz extracted from Romanian loess, the OSL growth curve as function of dose could be well fitted by a sum of two saturating exponential functions, and a marked difference between the saturation characteristics of fine and coarse grains was observed. This behaviour can be confidently attributed to quartz and not to feldspar contamination, as it was also observed in the case of pulsed-OSL stimulation where a deliberate rejection of any possible feldspar signal was achieved using signal gating. Photon- arrival-time distributions obtained in time resolved OSL investigations have been well fitted with one single exponential decay function, and the lifetimes obtained range in the typical interval for quartz ( $36.5\text{--}38.5\mu\text{s}$ ) for natural as well as regenerated signals for doses as high as 1 kGy. The lack of dependency of luminescence lifetimes with dose strongly suggests that there is only one luminescence centre emitting in our detection window. Thus, the second saturating exponential component observed in the dose response pattern cannot be interpreted as the result of the presence of another UV emitting centre, although we cannot rule out competition with another center which is either non-radiative or emitting in a different detection window. Nonetheless, the results of the present study clearly indicate that the age discrepancy obtained on fine and coarse grains of loess is more widespread than previously thought; it is not restricted to just the Lower Danube basin but applies to loess from Vojvodina as well. In the absence of a clear understanding of the causal mechanism, we recommend caution with interpretation of the quartz SAR-OSL ages for samples that display a double exponential dose response curve, at least in the high dose range.

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## Table captions

**Table 1.** Equivalent dose estimation results. The uncertainties mentioned with luminescence and radionuclide specific activity data are random errors. Data marked by \* represent the results obtained applying a thermal treatment consisting of ph 10 s at 260 °C and cutheat to 180 °C. These additional experiments have been performed in order to increase the confidence in the robustness of the equivalent doses obtained and to further emphasize that these values are not affected by the thermal treatment applied. The IR stimulation treatment implied in the IR depletion ratio tests (**Duller, 2003**) consisted of 40 s stimulation at a temperature of 60°C.

**Table 2.** Summary of the luminescence age results. The uncertainties coming along with the luminescence and the dosimetry data (including annual dose rates) are random; the uncertainties indicated with the optical ages are the overall uncertainties. All associated uncertainties represent 1  $\sigma$ . The associated uncertainties were calculated based on the error assessment system reported by **Aitken and Alldred (1972)** and **Aitken (1976)**. The quoted water content was determined based on the difference between the natural “as found” and the dry weight of the material extracted from the ends of the tubes, with a relative error of 25%. The total dose rate comprises the contribution from the alpha, beta and gamma radiations as well as the contribution of the cosmic rays; in the case of the coarse fraction a factor of 0.01 Gy/ka was adopted to account for the internal contribution (**Vandenbergh et al., 2008**). Cosmic ray contribution to the total dose rate was derived based on the equations published by **Prescott and Hutton (1994)**. The dose rates were calculated using the conversion factors tabulated by **Adamiec and Aitken (1998)**. For fine grains the alpha efficiency factor considered was  $0.04 \pm 0.02$ . A beta attenuation and etching factor of  $0.94 \pm 0.045$  was considered for 63-90  $\mu\text{m}$  grains (**Aitken 1985, Appendix C**).

## Figure captions

**Figure 1.** Study area. Map of Vojvodina (Northern Serbia) (a) with the Tamiš loess plateau indicated and enlarged (b) (Marković et al., 2014, modified).

**Figure 2.** Stratigraphic column, description, sediment colour and magnetic properties of the investigated section compared with MIS (Martinson et al, 1987) and luminescence age results. The location of the samples investigated by luminescence is indicated by arrows (Marković et al., 2014, modified).

**Figure 3.** Dose recovery results for fine (4-11  $\mu\text{m}$ ) grains (panel a) and coarse (63-90  $\mu\text{m}$ ) grains (panel b) (Marković et al. 2014). Natural aliquots were bleached twice for 100 s at room temperature using the blue light emitting diodes; the two bleaching treatments were separated by a 10 ks pause. The aliquots were then given a known dose chosen to be equal to the estimated equivalent dose, and measured using the SAR protocol. The solid line (eye guide) represents the 1:1 relation; the dotted lines (eye guide) bracket a 10% deviation from unity.

**Figure 4.** (a) Results of the equivalent dose preheat plateau test. (b) Results of the high resolution pulse anneal test for natural and regenerated signal. For natural signals each data point presents the average value obtained on three aliquots of the ratio between the natural signal measured after different preheat (10 s) temperatures and a 16 Gy test dose signal. In the case of regenerated signals three fresh aliquots have been bleached, given a dose approximately equal to the equivalent dose, the OSL signal being measured at 125°C following different preheat (10 s) temperatures, each value being normalized to the response to a constant test dose of 16 Gy (cutheat to 180 °). The insert presents data obtained using Risø calibration quartz.

**Figure 5.** Comparison of average SAR-OSL growth curves for four samples of different ages using fine (4-11  $\mu\text{m}$ ) grains and coarse (63-90  $\mu\text{m}$ ) grains. The number of aliquots (n) used to obtain the average is specified in the legend. The average dose response fitting function for all investigated aliquots of a certain grain size is also given.

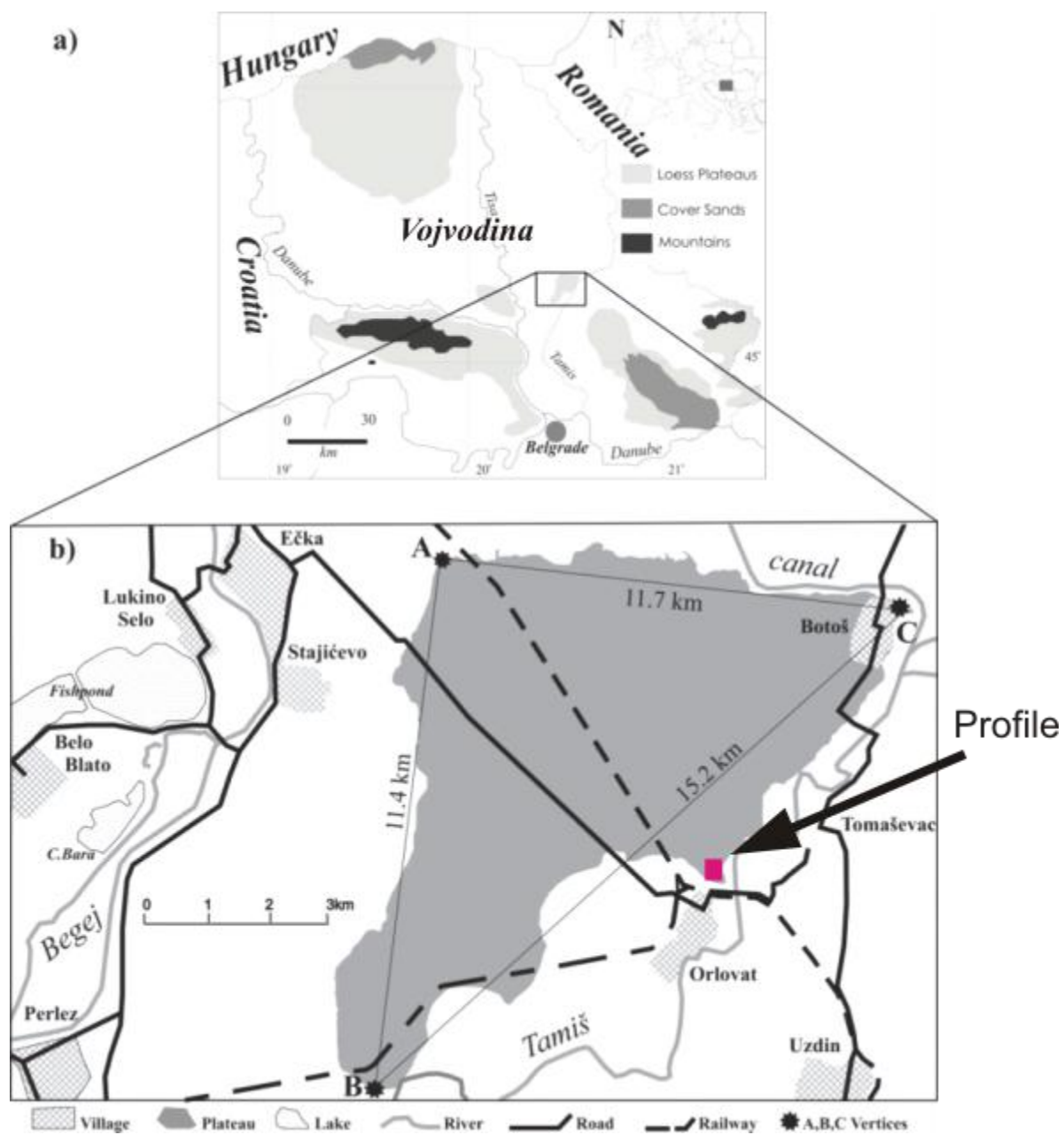
**Figure 6.** Photon arrival time distribution recorded during on and off time. The sample was preheated to 220 °C and stimulated at 125°C.

**Figure 7.** (a) Time resolved OSL curves (off time shown) of natural and regenerated signals of a representative aliquot of sample ORL 4 fine (4-11  $\mu\text{m}$ ) grains produced for an on time of 50  $\mu\text{s}$  and an off time of 450  $\mu\text{s}$ , by adding up signals recorded in 100s. Panel (b) presents the same

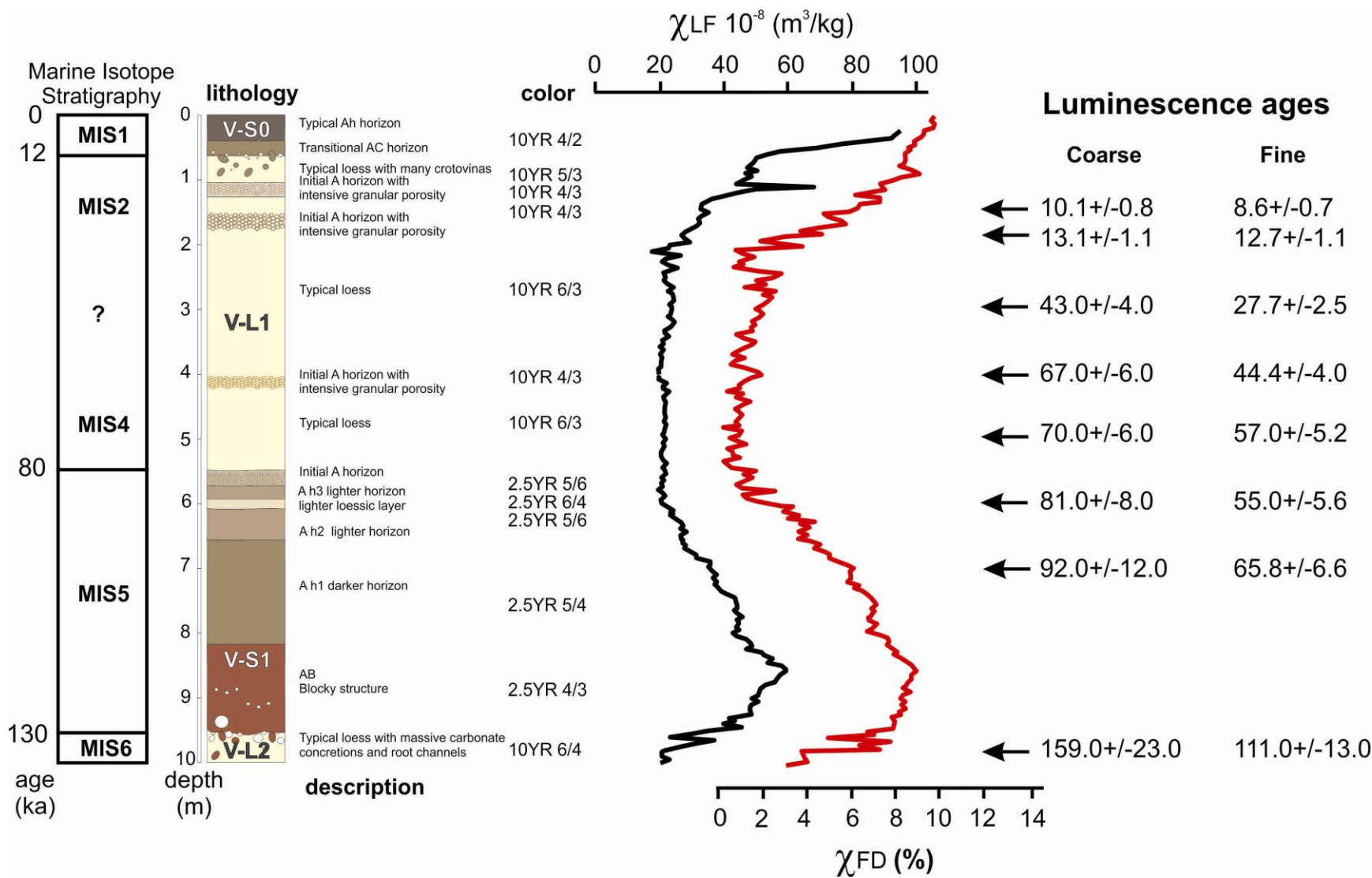
data enlarged for the first 50  $\mu$ s of the off time. The solid lines represent the best fit (single exponential decay plus a constant (background)) of the data. Please note the logarithmic y-axis.

**Figure 8. (a)** Dose response curves for fine (4-11  $\mu$ m) grains and coarse (63-90  $\mu$ m) grains of sample ORL 4 constructed using pulsed OSL data. Each data point represents the average value obtained on four aliquots. The aliquots have been measured using a typical SAR protocol measurement sequence. The test dose was 19 Gy. The photomultiplier was gated to record only during off time. Data was registered in 500 channels of 0.2 seconds, each channel consisting of 400 pulses of stimulation. Data collected from second 1 to second 4 of signal registration was selected for integration, which corresponds to a stimulation time interval of 0.32 seconds, a value very similar to that used in CW-OSL (0.308s). **(b)** Average lifetimes obtained as function of dose. The lifetimes of natural signals are depicted as stars. The IR stimulation was performed in a pseudo-CW stimulation manner (on time 500  $\mu$ s; off time 4.5  $\mu$ s) for 100 s at 125 °C.

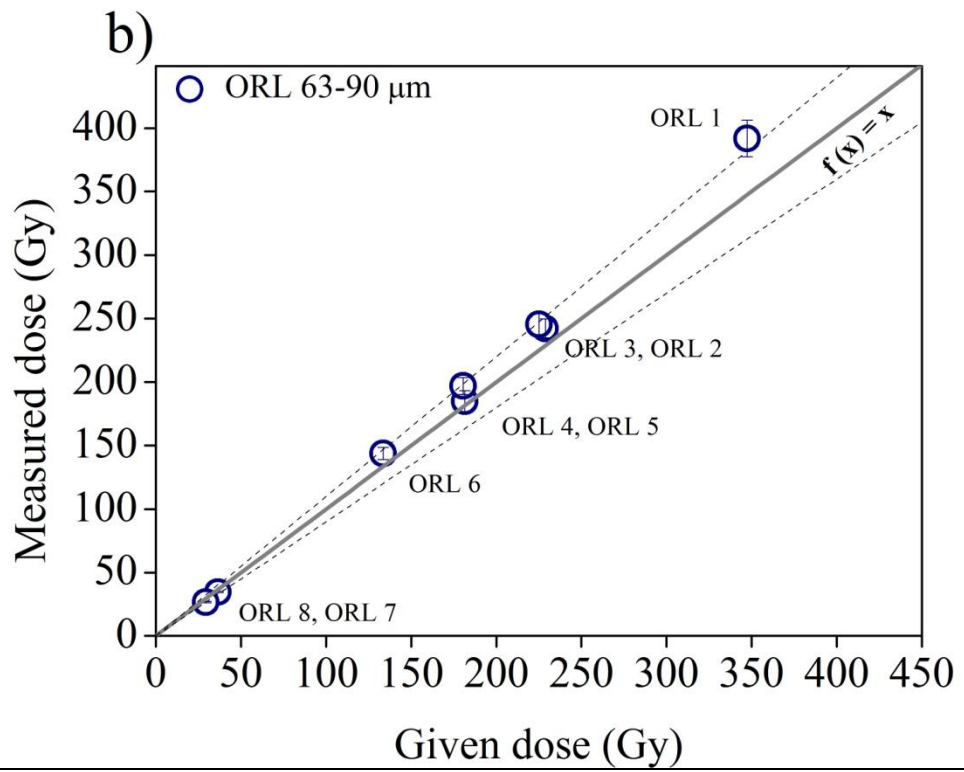
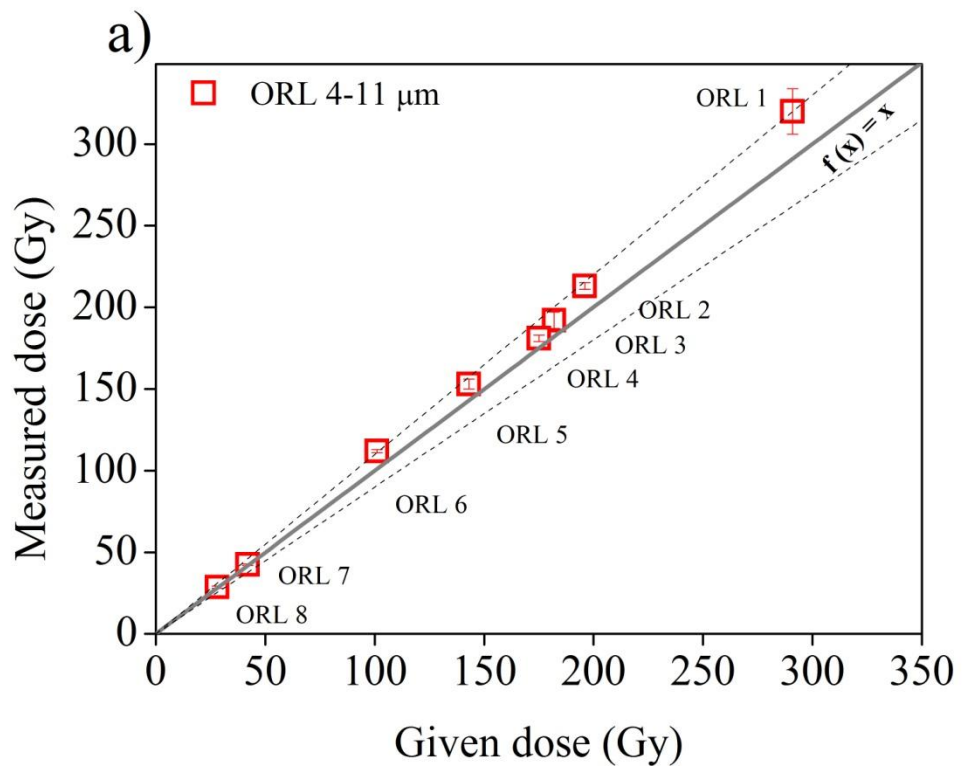
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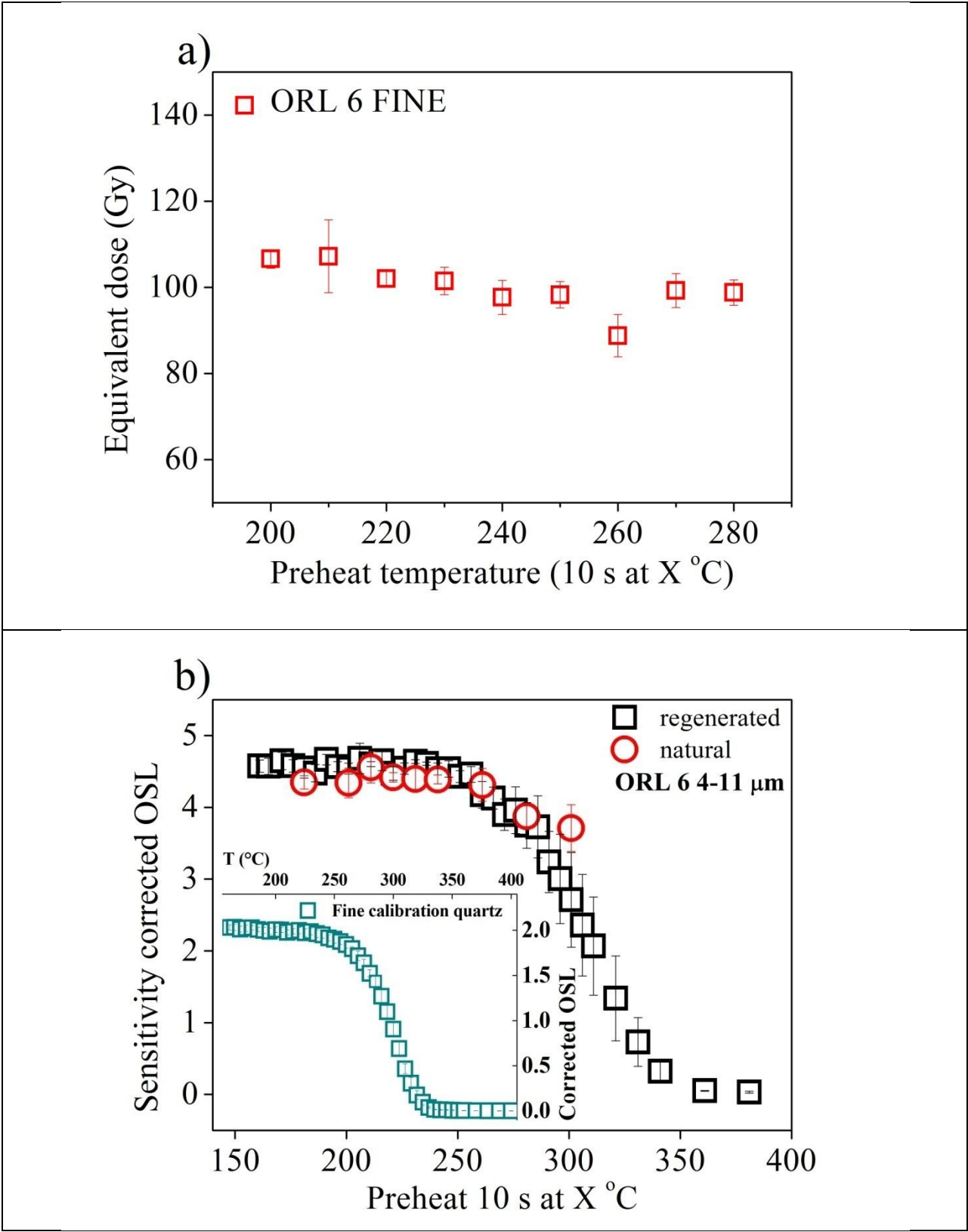
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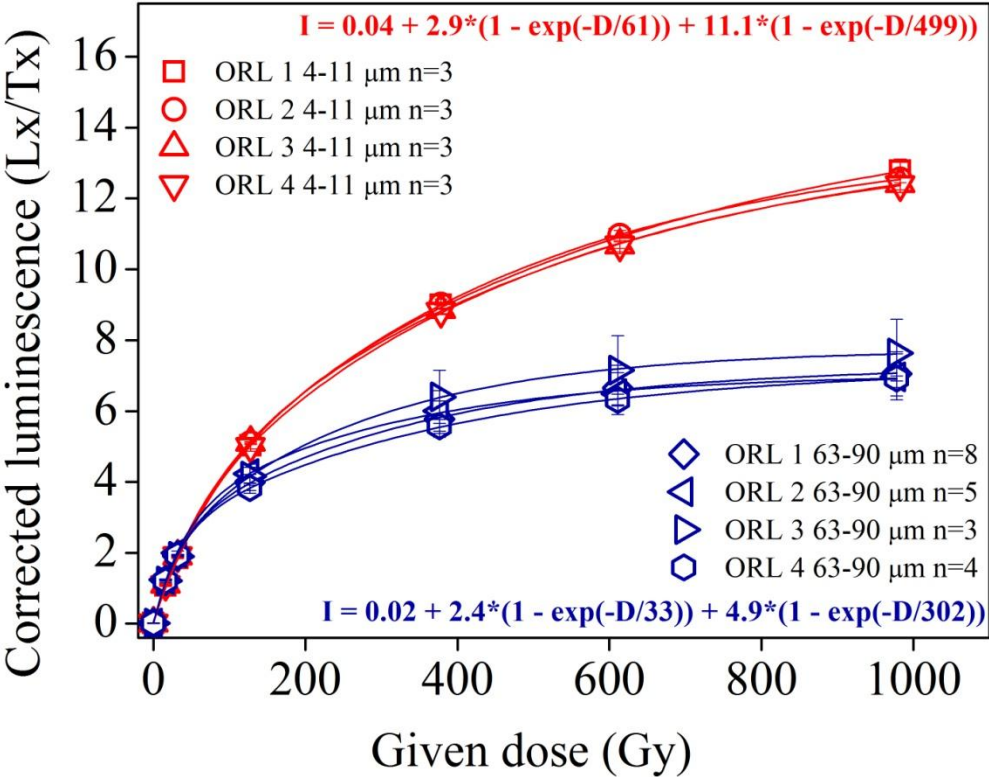


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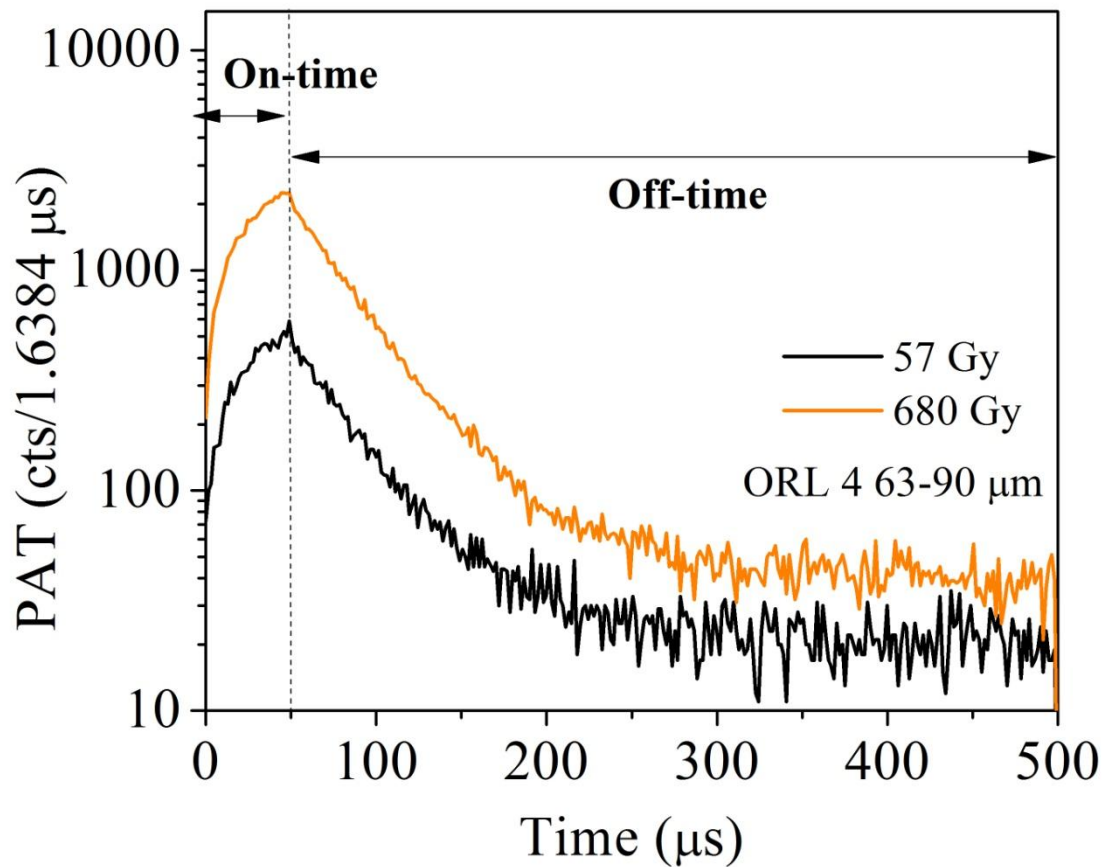




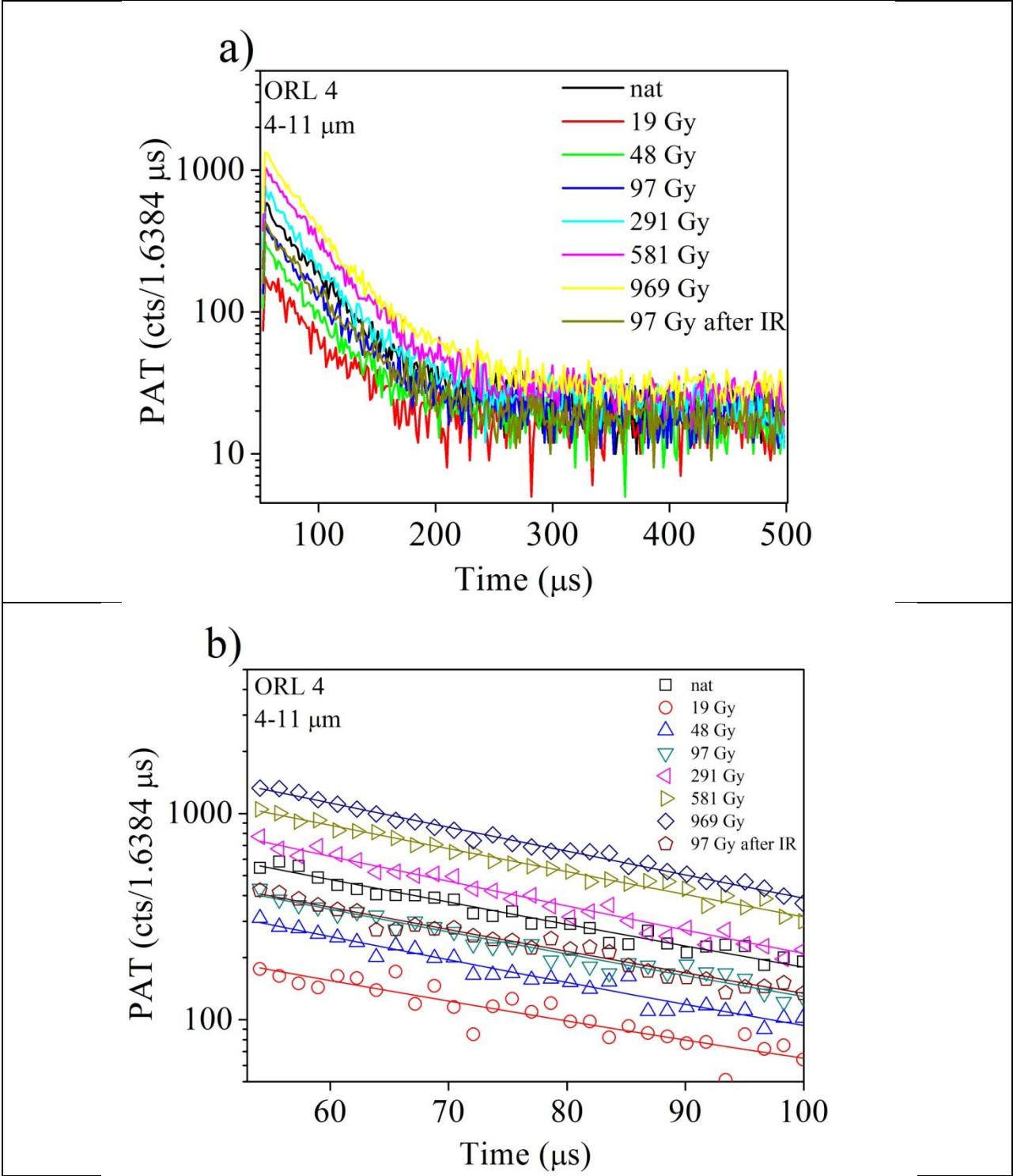
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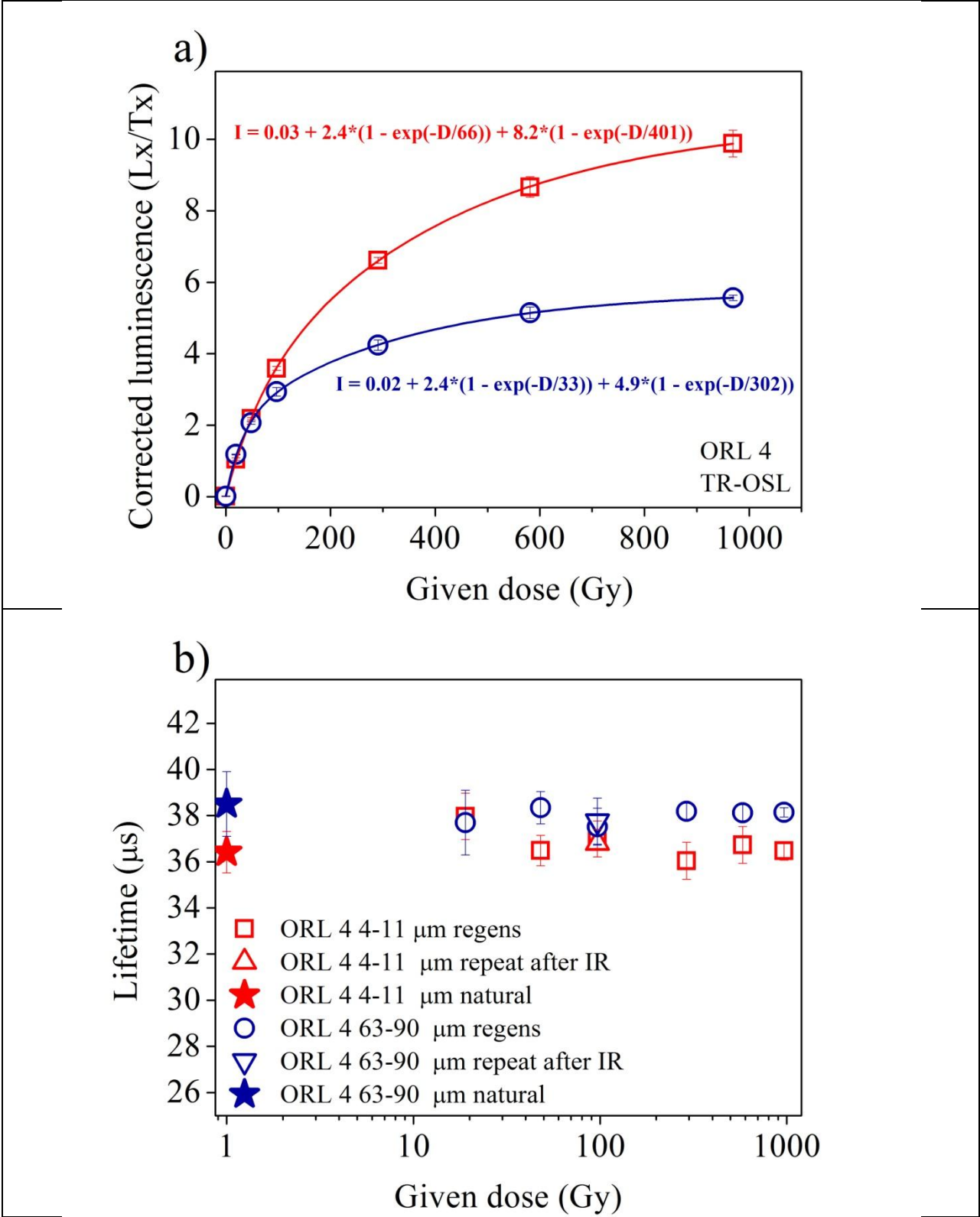
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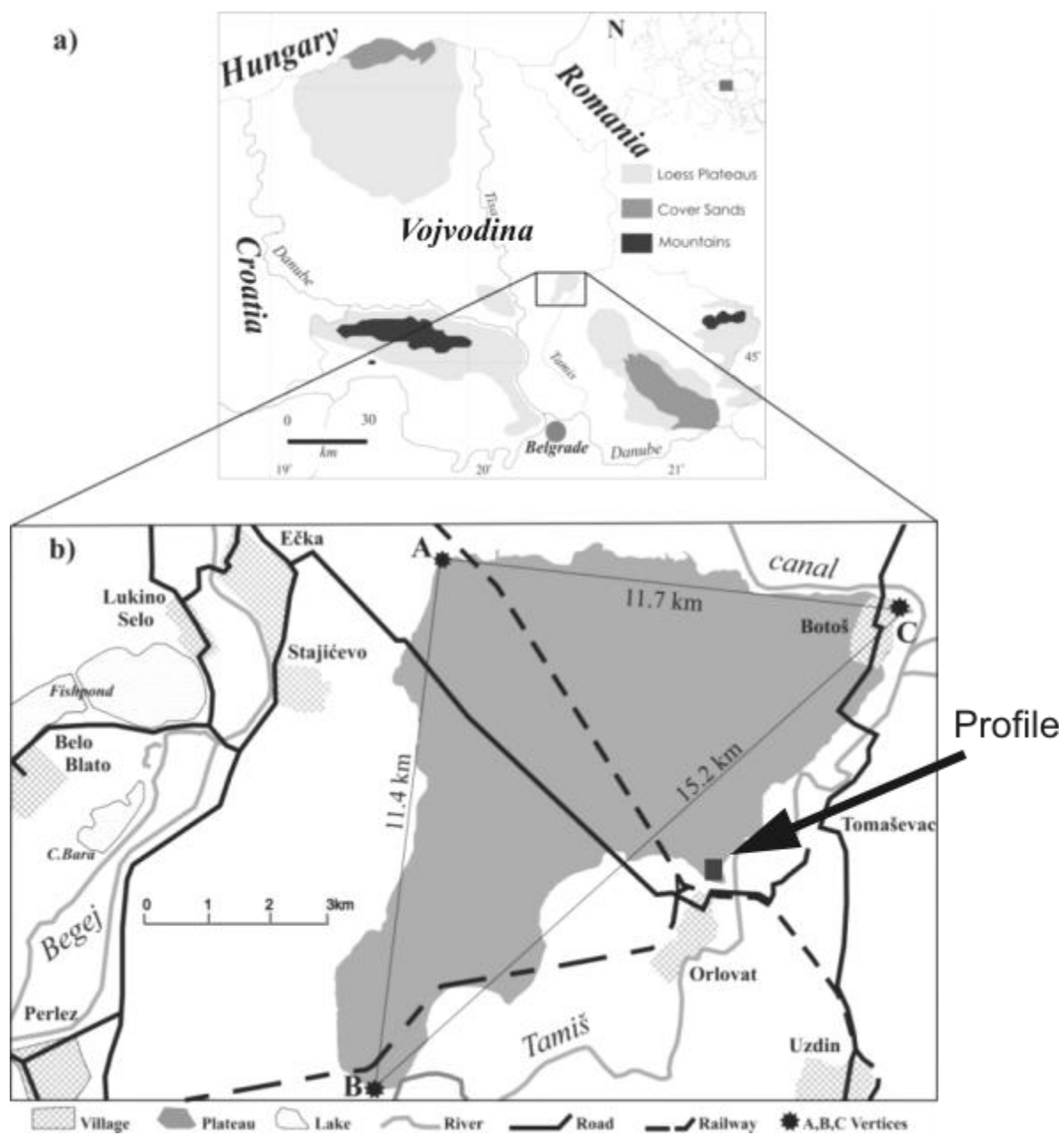
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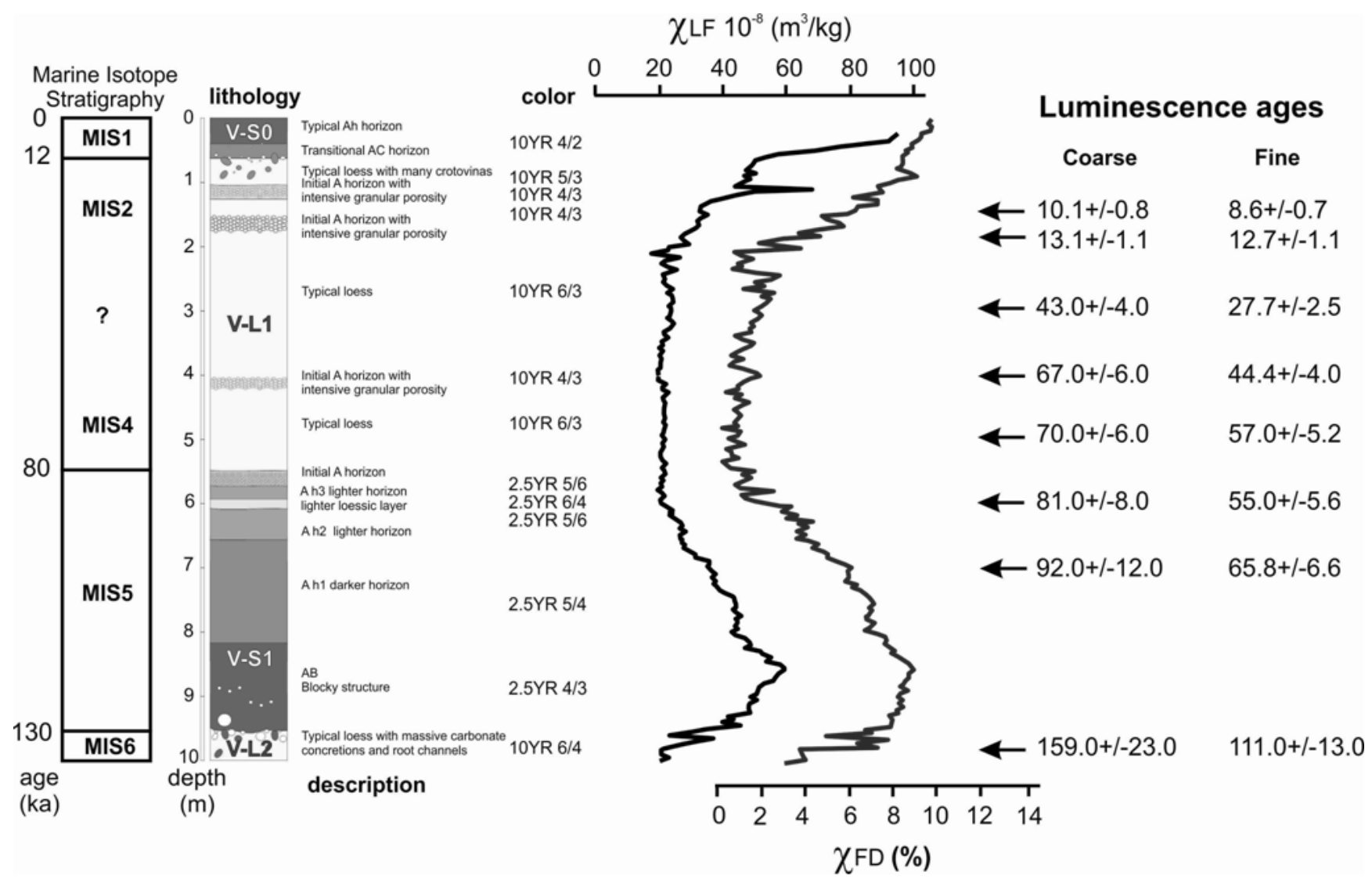
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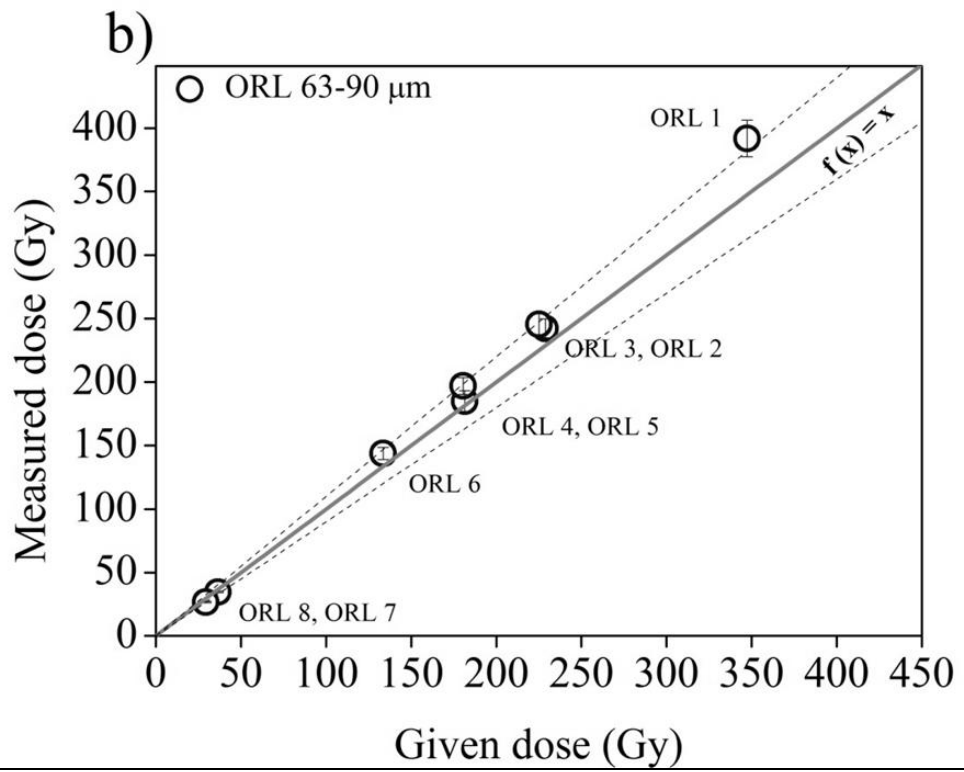
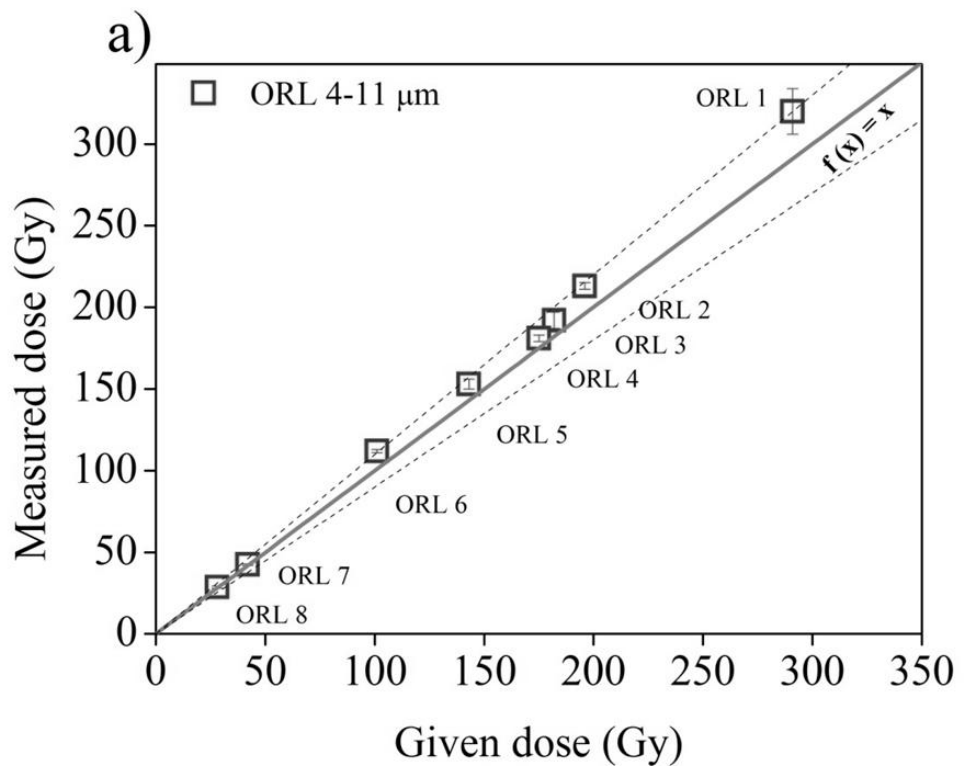
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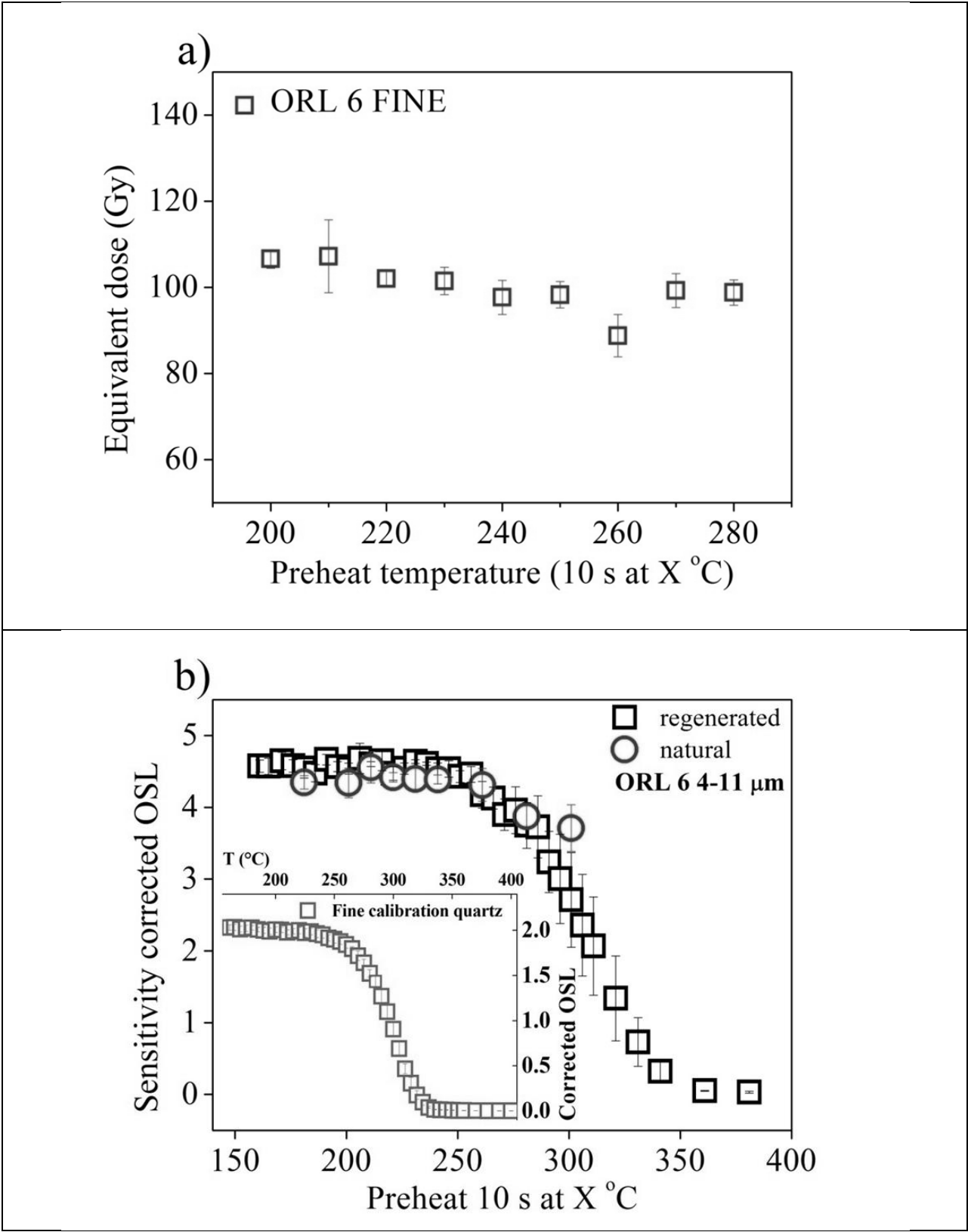
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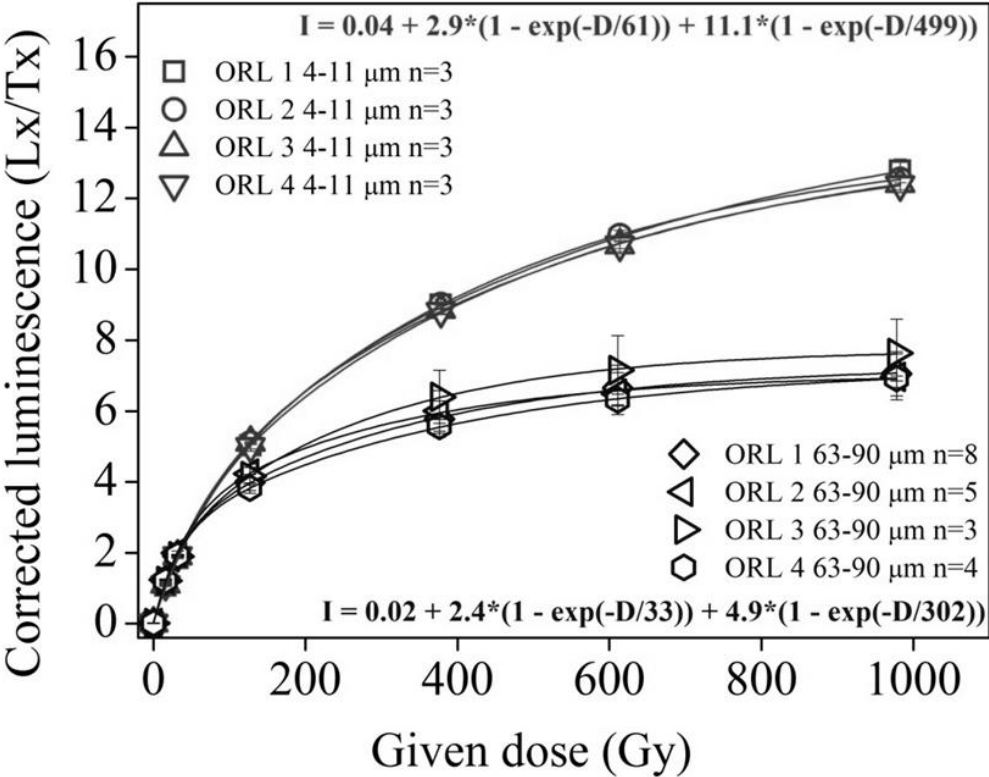


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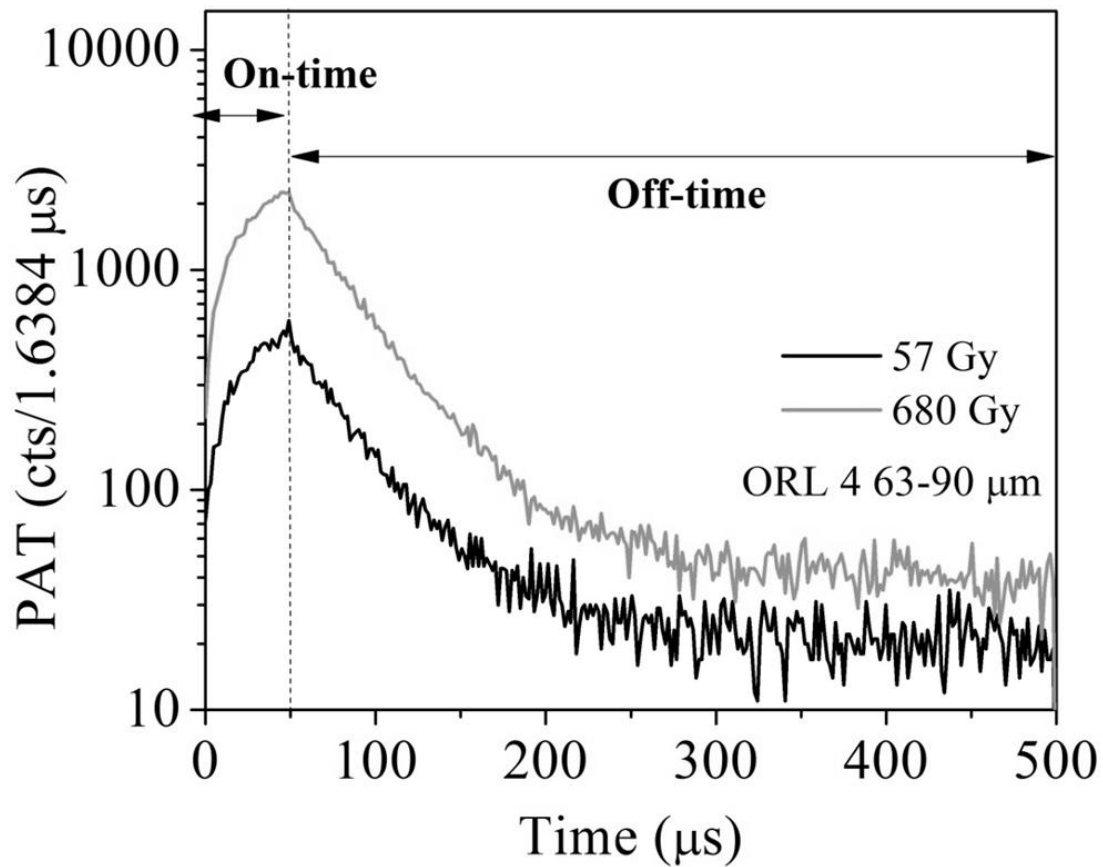




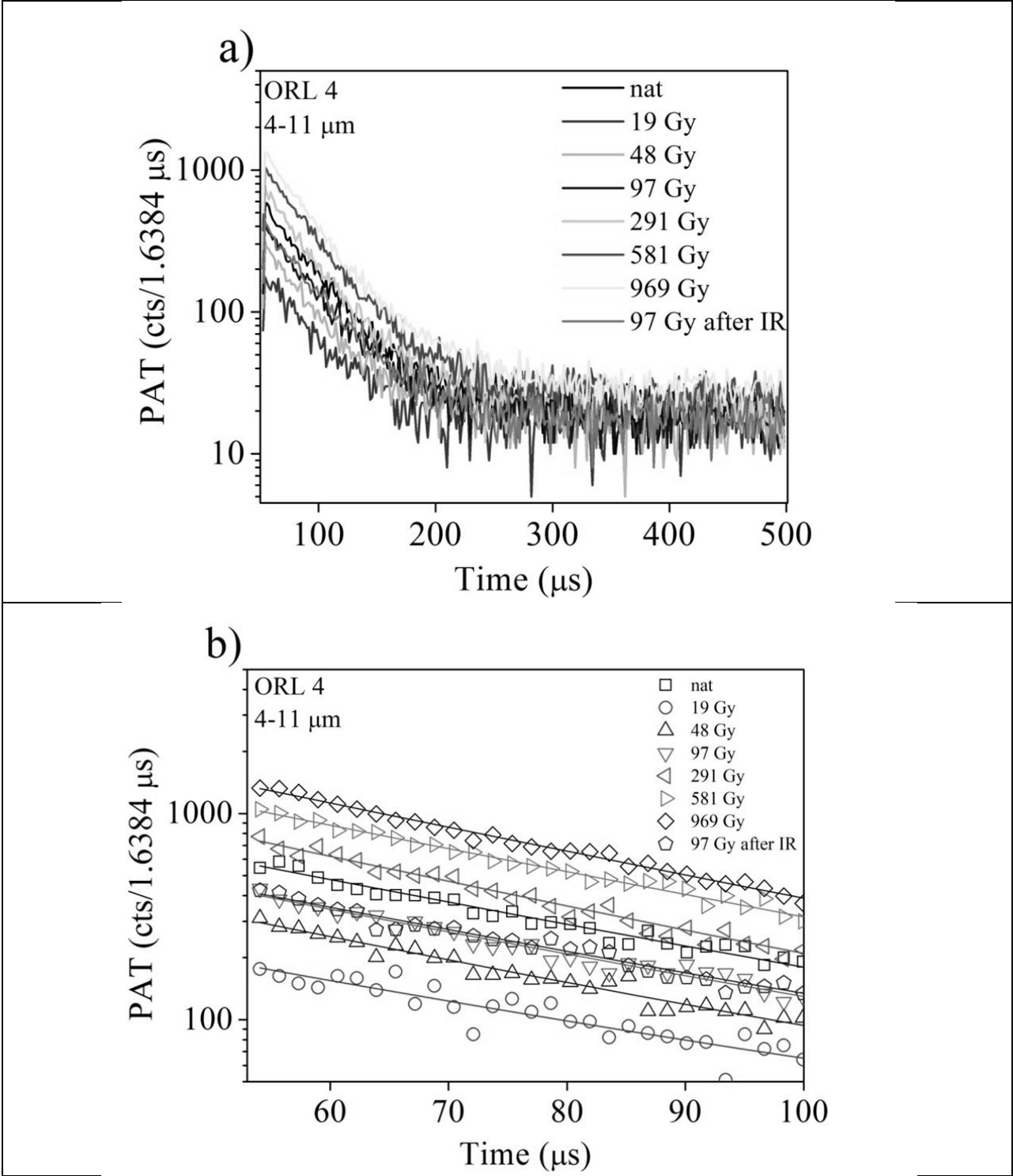
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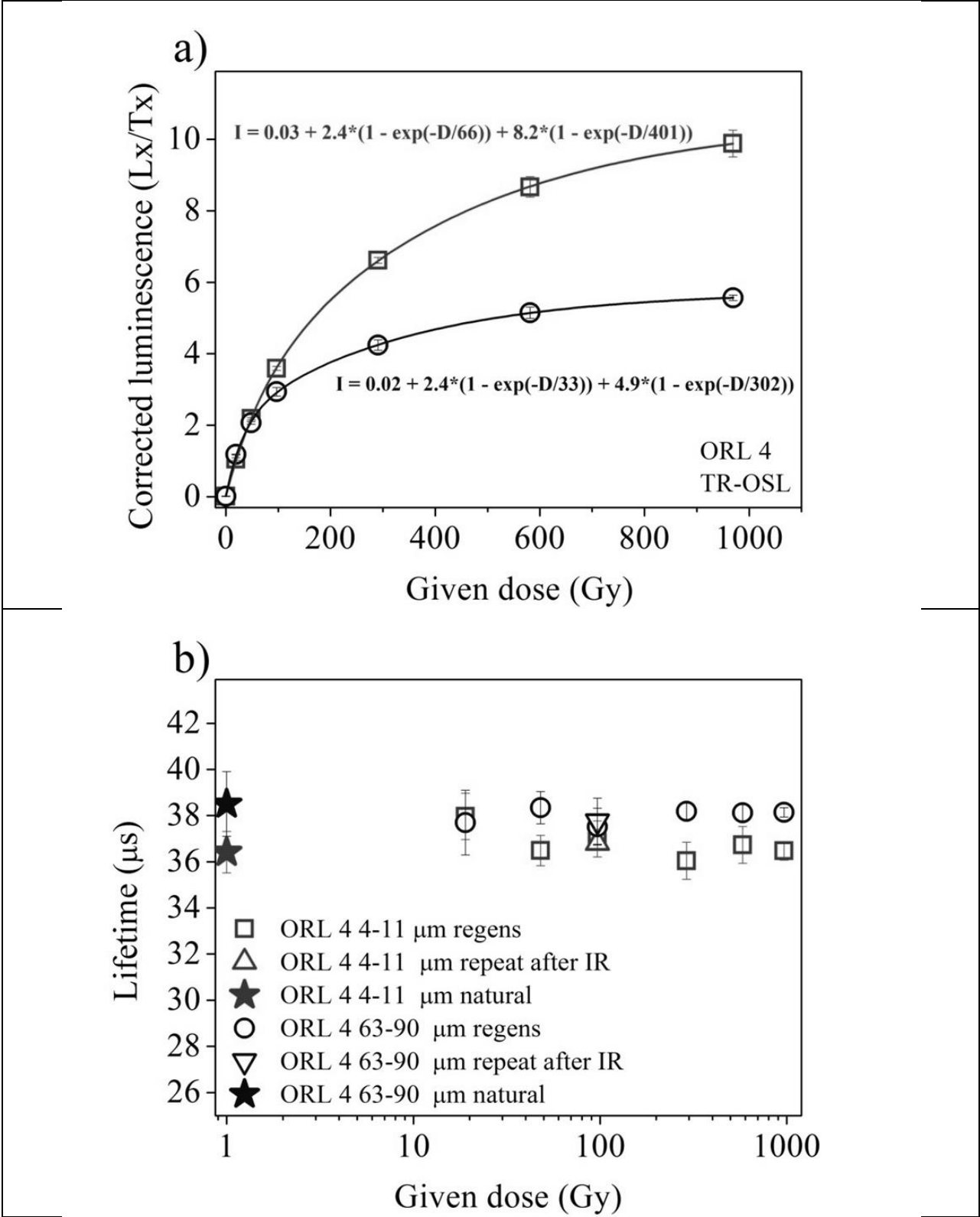
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Table

SAMPLE	Grain size	Number of aliq	Equivalent Dose (Gy)	Recycling ratio	IR depletion ratio	Recuperation (%)
ORL 1	4-11 $\mu\text{m}$	8/9	$292 \pm 10$	$0.96 \pm 0.01$	$0.96 \pm 0.01$	$0.09 \pm 0.01$
	63-90 $\mu\text{m}$	8/12	$347 \pm 38$	$0.91 \pm 0.01$	$0.93 \pm 0.01$	$0.19 \pm 0.03$
ORL 2	4-11 $\mu\text{m}$	9/9	$196 \pm 4$	$0.95 \pm 0.01$	$0.96 \pm 0.01$	$0.10 \pm 0.01$
	63-90 $\mu\text{m}$	5/8	$229 \pm 24$	$0.90 \pm 0.01$	$0.91 \pm 0.01$	$0.20 \pm 0.08$
ORL 3	4-11 $\mu\text{m}$	9/9	$182 \pm 4$	$0.95 \pm 0.01$	$0.97 \pm 0.01$	$0.10 \pm 0.02$
	63-90 $\mu\text{m}$	10/16	$225 \pm 13$	$0.94 \pm 0.01$	$0.94 \pm 0.01$	$0.09 \pm 0.02$
ORL 4	4-11 $\mu\text{m}$	8/9	$175 \pm 4$	$0.95 \pm 0.01$	$0.98 \pm 0.01$	$0.08 \pm 0.01$
	63-90 $\mu\text{m}$	11/16	$181 \pm 10$	$0.94 \pm 0.01$	$0.94 \pm 0.01$	$0.15 \pm 0.06$
ORL 5	4-11 $\mu\text{m}$	7/7	$143 \pm 3$	$0.97 \pm 0.01$	$0.98 \pm 0.01$	$0.02 \pm 0.02$
	4-11 $\mu\text{m}^*$	7/7*	$140 \pm 5$	$0.98 \pm 0.01$	$0.95 \pm 0.01$	$0.33 \pm 0.23$
	63-90 $\mu\text{m}$	10/14	$180 \pm 10$	$0.96 \pm 0.01$	$0.96 \pm 0.01$	$0.05 \pm 0.02$
ORL 6	4-11 $\mu\text{m}$	7/7	$102 \pm 2$	$0.98 \pm 0.01$	$0.96 \pm 0.01$	$0.01 \pm 0.04$
	4-11 $\mu\text{m}^*$	7/7*	$95 \pm 3$	$0.97 \pm 0.01$	$0.96 \pm 0.02$	$0.10 \pm 0.07$
	63-90 $\mu\text{m}$	13/13	$133 \pm 7$	$0.96 \pm 0.01$	$0.95 \pm 0.01$	$0.07 \pm 0.01$
ORL 7	4-11 $\mu\text{m}$	9/9	$42.0 \pm 0.5$	$1.01 \pm 0.01$	$1.00 \pm 0.01$	$0.07 \pm 0.03$
	4-11 $\mu\text{m}^*$	7/7*	$41.7 \pm 1.0$	$0.98 \pm 0.02$	$1.00 \pm 0.01$	$0.04 \pm 0.03$
	63-90 $\mu\text{m}$	13/13	$36.3 \pm 1.8$	$0.96 \pm 0.01$	$0.97 \pm 0.01$	$0.09 \pm 0.01$
ORL 8	4-11 $\mu\text{m}$	9/9	$28.2 \pm 0.3$	$1.00 \pm 0.01$	$0.99 \pm 0.01$	$0.09 \pm 0.03$
	4-11 $\mu\text{m}^*$	7/7*	$29.1 \pm 0.3$	$0.99 \pm 0.02$	$0.98 \pm 0.02$	$0.18 \pm 0.02$
	63-90 $\mu\text{m}$	13/13	$28.8 \pm 1.1$	$0.95 \pm 0.01$	$0.97 \pm 0.01$	$0.11 \pm 0.01$

Table

SAMPLE (4-11 μm)	Grain size	Depth (cm)	Equivalent dose (Gy)	Ra-226 (Bq/kg)	Th-232 (Bq/kg)	K-40 (Bq/kg)	Water Content (%)	Anual Dose (Gy/Ka)	Age (Ka) Total error	Random Error ( %)	Systematic error ( %)
ORL 1	4-11 μm	965	292 ± 10	32.4 ± 0.9	35.6 ± 0.3	406 ± 4	20	2.63 ± 0.03	111 ± 13	3.6	11.1
	63-90 μm		347 ± 38					2.18 ± 0.02	159 ± 23	11	9.2
ORL 2	4-11 μm	700	196 ± 4	31.9 ± 0.6	38.6 ± 0.4	456 ± 7	15	2.98 ± 0.04	65.8 ± 6.6	2.4	9.8
	63-90 μm		229 ± 24					2.49 ± 0.02	91.9 ± 12.1	10.5	7.9
ORL 3	4-11 μm	600	182 ± 4	37.0 ± 0.6	38.2 ± 0.3	471 ± 7	10	3.31 ± 0.04	55.0 ± 5.6	2.5	9.9
	63-90 μm		225 ± 13					2.76 ± 0.02	81.4 ± 8.0	5.8	7.9
ORL 4	4-11 μm	500	175 ± 4	32.1 ± 0.4	36.0 ± 0.4	444 ± 7	10	3.07 ± 0.04	57.0 ± 5.2	2.6	8.8
	63-90 μm		181 ± 10					2.57 ± 0.02	70.4 ± 6.1	5.6	6.7
ORL 5	4-11 μm	400	143 ± 3	34.4 ± 0.4	37.1 ± 0.3	462 ± 6	10	3.22 ± 0.04	44.4 ± 4.0	2.4	8.8
	63-90 μm		180 ± 10					2.70 ± 0.02	66.7 ± 5.8	5.6	6.7
ORL 6	4-11 μm	300	102 ± 2	37.4 ± 0.5	42.6 ± 0.3	546 ± 7	10	3.69 ± 0.04	27.7 ± 2.5	2.3	8.7
	63-90 μm		133 ± 7					3.10 ± 0.02	43.0 ± 3.7	5.3	6.7
ORL 7	4-11 μm	180	42.0 ± 0.5	35.7 ± 0.6	34.9 ± 0.4	481 ± 7	10	3.39 ± 0.04	12.7 ± 1.1	1.7	8.6
	63-90 μm		36.3 ± 1.8					2.78 ± 0.02	13.1 ± 1.1	5.0	6.6
ORL 8	4-11 μm	150	28.2 ± 0.3	34.8 ± 0.7	35.7 ± 0.2	476 ± 6	10	3.29 ± 0.04	8.6 ± 0.7	1.6	8.6
	63-90 μm		28.8 ± 1.1					2.77 ± 0.02	10.4 ± 0.8	3.9	6.6